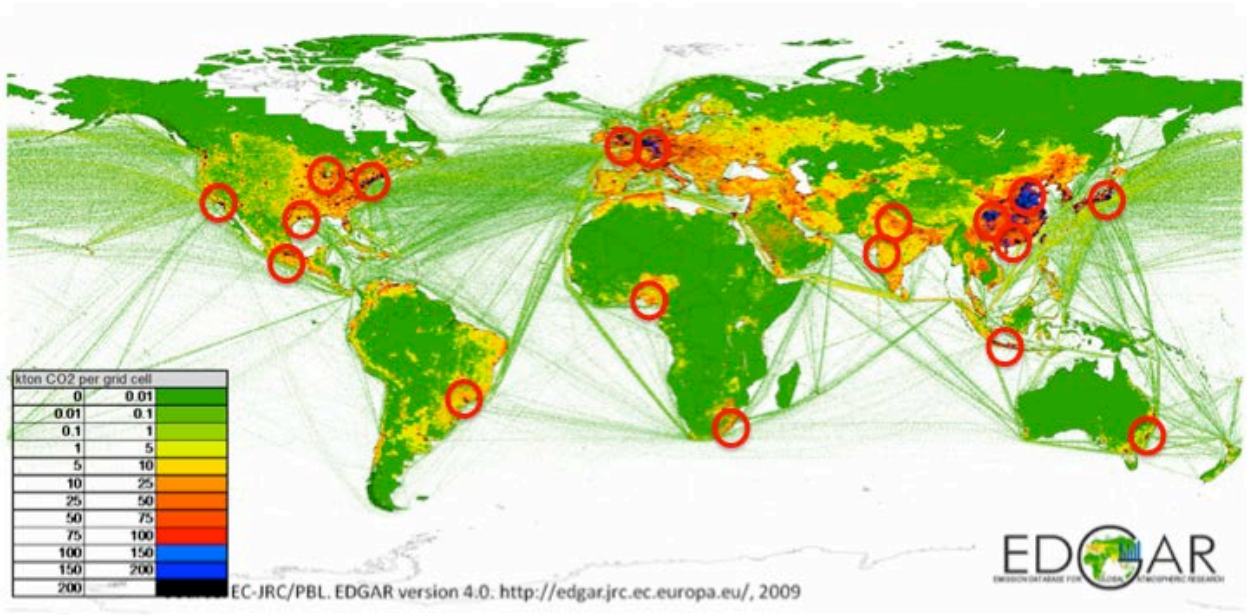

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http://www.kiss.caltech.edu/workshops/carbon2009/index.html

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Preamble: The problem and context

Can top-down estimates of carbon dioxide (CO_2) fluxes resolve the anthropogenic emissions of China, India, the United States, and the European Union with an accuracy of ±10% or better?

The workshop "Monitoring Exchange of Carbon Dioxide" was convened at the Keck Institute for Space Studies in Pasadena, California in February 2010 to address this question. The Workshop brought together an international, interdisciplinary group of 24 experts in carbon cycle science, remote sensing, emissions inventory estimation, and inverse modeling. The participants reviewed the potential of space-based and sub-orbital observational and modeling approaches to monitor anthropogenic CO_2 emissions in the presence of much larger natural fluxes from the exchange of CO_2 between the land, atmosphere, and ocean. This particular challenge was motivated in part by the NRC Report "Verifying Greenhouse Gas Emissions" [Pacala et al., 2010]. This workshop report includes several recommendations for improvements to observing strategies and modeling frameworks for optimal and cost-effective monitoring of carbon exchange.

The basic framing of the atmospheric CO_2 problem is captured in the 2007 IPCC Summary for Policy Makers [IPCC, 2007]:

> ***Global atmospheric concentrations of carbon dioxide, methane and nitrous oxide have increased markedly as a result of human activities since 1750 and now far exceed pre-industrial values. The global increases in carbon dioxide concentration are due primarily to fossil fuel use and land-use change.***
>
> *Carbon dioxide is the most important anthropogenic greenhouse gas (see Figure SPM-2). The global atmospheric concentration of carbon dioxide has increased from a pre-industrial value of about 280 ppm to 379 ppm[3] in 2005. The atmospheric concentration of carbon dioxide in 2005 exceeds by far the natural range over the last 650,000 years (180 to 300 ppm) as determined from ice cores. The annual carbon dioxide concentration growth-rate was larger during the last 10 years (1995 – 2005 average: 1.9 ppm per year), than it has been since the beginning of continuous direct atmospheric measurements (1960 – 2005 average: 1.4 ppm per year).*
>
> *The primary source of the increased atmospheric concentration of carbon dioxide since the pre-industrial period results from fossil fuel use, with land use change providing another significant but smaller contribution. Annual fossil carbon dioxide emissions[4] increased from an average of 6.4 [6.0 to 6.8] GtC (23.5 [22.0 to 25.0] $GtCO_2$) per year in the 1990s, to 7.2 [6.9 to 7.5] GtC (26.4 [25.3 to 27.5] $GtCO_2$) per year in 2000–2005. Carbon dioxide emissions associated with land-use change are estimated to be 1.6 [0.5 to 2.7] GtC (5.9 [1.8 to 9.9] $GtCO_2$) per year over the 1990s, although these estimates have a large uncertainty.*

Beginning with United Nations Framework Convention on Climate Change (UNFCCC, 1994), multilateral agreements to control the global emissions of greenhouse gases (GHG) have been proposed. A significant challenge that has hindered agreements to control GHG emissions has been the lack of a sufficiently accurate and transparent method to evaluate the actions of individual parties beyond self-reporting. The scientific community has decades of research on the carbon cycle and related fields that can be brought to bear in this important area of public policy. This report documents some of the ongoing and planned scientific research discussed at the workshop that can contribute to the development of transparent and rigorous methods to monitor anthropogenic CO_2 emissions with the accuracy required to support policy verification and assessment.

Attribution of emissions to specific natural or anthropogenic sources is a necessary component of resolving fossil fuel emissions of individual geographic regions, such as countries. According to the IPCC *Climate Change 2007: Synthesis Report*, CO_2 from fossil fuel use accounted for 56.6% of global anthropogenic GHG emissions (in terms of $C0_2$-equivalency), while anthropogenic changes to land use, such as deforestation, contribute another 17.3%; taken together, fossil fuel and land use account for over 70% of global anthropogenic CO_2 emissions. A few countries are responsible for the majority of the fossil fuel CO2 emission; China, India, the US, and the European Union account for approximately 44% of the national greenhouse gas emissions from all IPCC sectors of the top 20 emitters in 2000 (Pacala, 2010).

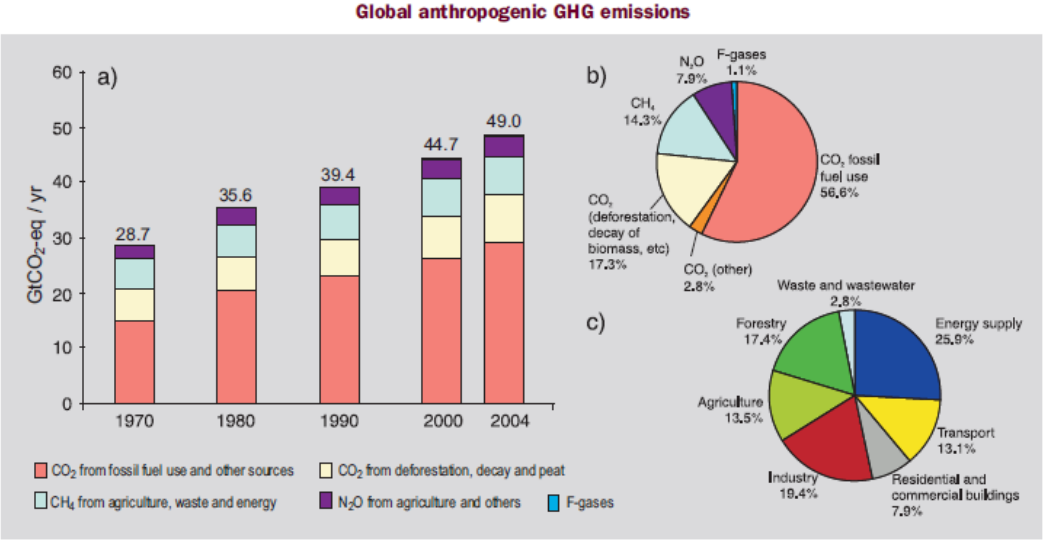

Figure 2.1. *(a) Global annual emissions of anthropogenic GHGs from 1970 to 2004.[5] (b) Share of different anthropogenic GHGs in total emissions in 2004 in terms of CO_2-eq. (c) Share of different sectors in total anthropogenic GHG emissions in 2004 in terms of CO_2-eq. (Forestry includes deforestation.) {WGIII Figures TS.1a, TS.1b, TS.2b}*

Estimating CO_2 emissions from fossil fuel combustion is typically performed using bottom-up inventory methods and top-down inverse calculations from atmospheric observations. Inventory accounting methods employ known sales of fossil fuels and economic sector activity data to estimate emissions. The IPCC has published standard protocols for calculating National Emissions Inventories (NEIs), and it is these estimates that are reported annually by signatories to the UNFCCC. However, the NEI protocols knowingly omit certain sources from reporting and uncertainties are calculated based only on published accounting practices. Top-down methods use observational data to produce objective estimates emissions and their associated uncertainties. These methods capture the total net exchange of CO_2 between the surface and atmosphere, but are plagued by inadequate observational sampling in space and time. Because they sample the total signal, it is difficult to provide confident attribution of emissions from specific natural or anthropogenic sources. Additionally, there are significant deficiencies in the atmospheric transport models that are required to infer flux estimates from atmospheric CO_2 concentration observations. There are few direct comparisons of top-down and bottom-up emissions estimates and reconciling such estimates for eg NEIs is necessary for effective long-term GHG emissions policy design, implementation, verification and assessment.

The current uncertainties in the science of carbon exchange, combined with limitations of current methods for attribution of the anthropogenic component, make the goal of resolving the fossil fuel emissions of specified countries to $\pm$10% accuracy a challenge that is currently unachievable. Participants in the workshop concluded that improvements in space-based and sub-orbital observing systems, combined with improvements to modeling frameworks, could make this goal achievable within a decade if an aggressive research and engineering agenda was pursued. This report contains specific suggestions for such improvements, including increased development of space-based observational capabilities, which is anticipated to play a greater role in both science and policy approaches to carbon monitoring.

The improvements in understanding and monitoring CO_2 exchange go beyond research science to have relevance to political agreements such as the international follow-on agreement to the Kyoto Accord and the United National Collaborative Program on Reducing Emissions from Deforestation and Forest Degradation in Developing Countries (REDD). International consensus on accurate, robust, and transparent methods for evaluating CO_2 flux becomes especially important. Carbon flux monitoring provides an unparalleled opportunity for research science to contribute to one of the most pressing problems of the present day and age.

Bottom-up Inventories

General overview of methods

The national greenhouse gas emissions inventories (NEIs) reported today are determined almost exclusively through the compilation of economic activity data that is converted to greenhouse gas emissions through the use of predetermined emissions factors in accordance with IPCC guidelines. This bottom-up approach is the foundation of the Measurement-Reporting-Verification (MRV) mechanism used at national and international levels for tracking GHG emissions. However, this process relies on spreadsheets, self-reporting, verification of the accounting procedures; it lacks any objective feedback from direct measurements of GHG emissions or other independent, scientifically validated observational data.

In 1992, United Nations Framework Convention on Climate Change (UNFCCC) signatories agreed "to develop, update periodically, publish and make available to the Conference of the Parties (COP) their national inventories of anthropogenic emissions by sources and removals by sinks, of all GHG not controlled by the Montreal Protocol [and] to use comparable methodologies for inventories of GHG emissions and removals, to be agreed upon by the COP (IPCC 1996)." The *Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories* were developed to provide instructions for such emissions calculations for the signatories of the UNFCCC. This document develops internationally-agreed upon methodology and software for calculation and reporting of national GHG emissions and establishes a data management system for collection, review and reporting of national data. Importantly, the IPCC guidelines intended to create "a common and consistent mechanism that enables Parties to the UNFCCC to compare the relative contribution of different emission sources and greenhouse gases to climate change (EPA 2010)."

In the US, the US Environmental Protection Agency (EPA) is the federal agency responsible for preparing the UNFCCC greenhouse gas emissions estimates. These estimates were most recently presented in the 2010 report, *Inventory of U.S. Greenhouse Gas Emissions and Sinks: 1990 – 2008*. According to the report, estimates were calculated using methodologies consistent with the recommendation of the *Revised 1996 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC/UNEP/OECD/IEA 1997), the *IPCC Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories* (IPCC 2000), and the *IPCC Good Practice Guidance for Land Use, Land-Use Change, and Forestry* (IPCC 2003). EPA has begun to incorporate methodology and data from the *2006 IPCC Guidelines for National Greenhouse Gas Inventories* (IPCC 2006). The report is consistent with the UNFCCC guidelines for inventory reporting.

EPA uses a decentralized approach in preparing the US annual inventory. Multiple federal and state government agencies, research and academic institutions, industry associations, and private consultants supply data to EPA for preparation of the report, or are involved in reviewing and otherwise preparing

the report. The EPA's Office of Atmospheric Programs has primary responsibility for preparation of the report, which is officially submitted to the UNFCCC by the US Department of State. Other government agencies that formally contribute official data for use in the Inventory include the US Department of Energy's Energy Information Administration and the US Department of Defense, while other federal agencies provide activity data for use in EPA's emissions calculations through informal relationships.

The National Inventory Report (NIR) includes emissions calculations carried out in a standardized and specified manner, Common Reporting Format (CRF) tables, and a 'verification and uncertainty assessment.' CRF tables specify standard table format using common source/sink categories, common fuel categories, and common definitions of pollutants, units, and time intervals. 'Verification' refers to a specific set of simple checks for completeness and accuracy of submissions that must be followed. Individual source leads calculate emissions and sink estimates and develop relevant report text, sectoral background tables for the CRF, and accompanying quality assurance, quality control, and uncertainty analysis. As much as possible, the report relies on published activity and emission factor data. Activity data varies by emission source category and includes information such as fuel consumption or deliveries, vehicle-miles traveled, or raw material processed. Emission factors are used to relate activity data to quantities of emissions.

National Emissions Inventory Precision and Uncertainty

Uncertainty information is associated with inventory emissions estimates. The 1996 IPCC Guidelines (IPCC/UNEP/OECD/IEA 1997) require that countries provide single point estimates for each gas and emission or removal source category. The US inventory overall uncertainty estimate was developed using the IPCC Tier 2 uncertainty estimation methodology. Specific factors affect the uncertainty associated with each estimate; some estimates are well known and have low uncertainty, while other estimates have uncertainties that are not well understood or well characterized. The US EPA report recommends additional research in specific areas for help reduce uncertainty in the US inventory: incorporation of excluded emissions sources, improvement in the accuracy of emissions factors used to calculate emissions quantities from activity data, and collection of additionally detailed activity data. The US inventory and current IPCC direction for uncertainty quantification are focused entirely on the uncertainty assigned to parts of the calculation of emissions from activity data and emissions factors; no discussion of independent validation of these quantities is included in uncertainty discussions.

Beyond the assignment of uncertainty to emissions values in inventory reports per IPCC guidelines, uncertainty in CO_2 emissions can be estimated in at least four different ways [Marland et al., 2008]:

1. Comparison of estimates made by independent methods
2. Comparison of estimates from multiple sources
3. Evolution over time of estimates from a single source
4. Modeling against remotely sensed data

Two other aspects of uncertainty in reported emissions are the independence of activity data used in emissions calculations and the trend of uncertainty in the overall global emissions estimates. First,

estimates of CO_2 from different sources are rarely truly independent, as they often use the same sources of data on fuel consumption [Marland et al., 2008]. However, the subsequent calculations are still useful comparisons, as the differences in methodologies and values used in the calculations are often not as standardized and therefore provide opportunities for development of best practices. Finally, global contributions to greenhouse gas concentrations from countries with larger uncertainties are increasing. This has the effect of increasing the overall uncertainty in global emissions estimates, further underscoring the need for improved methodologies to reduce uncertainty in emissions estimates.

Lateral transport

National inventories of greenhouse gas emissions prepared according to UNFCCC guidelines have other limitations. One important limitation is the exclusion of lateral transport in the UNFCCC methodology. The UNFCCC accounting methodology accounts for emissions produced within a given region or nation; however, a portion of fossil fuel emissions are associated with the production of goods that are exported (laterally transported) from a given country and consumed in a different, often more developed, country. The producing country is held accountable for the emissions associated with these goods, which can be considered to be a contribution to the disparity in emissions between high-import, developed countries, and manufacturing-intensive, high-export, less developed countries [Davis et al., 2009]. An analysis of country-based emissions inventories that accounts for emissions associated with imports and exports by assigning the emissions of productions of an amount of goods to the country that ultimately consumes the goods paints a different picture of per country and per capita emissions. Adoption of an accounting methodology that accounts for such 'carbon leakage' due to international trade may help facilitate progress on international emissions reductions agreements that are currently stumbling over disagreements of responsibility for regional and historical emissions. Current UNFCCC guidelines specifically exclude accounting for carbon leakage, stating that "In line with the principle of national emissions, the IPCC methodology accounts for the bulk of greenhouse gas emissions related to fuel combustion in the country in which those emissions are released (IPCC 2006)." Failure to include these emissions leads to known inaccuracies in the total estimate of global anthropogenic GHG emissions. We recommend that, at a minimum, estimates of total emissions due to lateral transport and a quantified uncertainty in these emissions be included in NEI accounting to enable an accurate global GHG emissions budget.

Other Emissions Inventories

In addition to the NEIs reported to the UNFCCC, there are a number of emissions inventories that have been developed by the research community. The leading research emissions inventories (CDIAC [Andres et al., 2010], EDGAR [EDGAR, 2009], Vulcan [Gurney et al., 2009], FFDAS [Rayner et al., 2010], etc.) have limitations in the accuracy of their absolute CO_2 emissions at the national/annual level, as well as in the spatial and temporal redistribution of emissions over smaller space and time scales. Each inventory applies different approaches for the spatial distribution of CO_2 emissions based on proxies such as population distribution, space-based observations of night-lights, power plant fuel consumption

statistics, etc.; however each of these proxies involves a number of assumptions which impart spatial biases on the inventory, producing a significant source of potential error during attribution of the emissions to smaller scales like economic/industrial sectors, states/cities or smaller point sources (factories or power plants). For example, spatial attribution of CO_2 emissions by night-lights assumes that the integrated 24-hour emissions are proportional to those observed during the nightlights overpasses (typically 10:30 pm local time). The use of population distributions to distribute emissions will also induce bias, along with the issue that some industrial activities that may generate high levels of CO_2 emissions (extraction of petroleum from oil sands) can be located hundreds of kilometers away from major population centers. Furthermore, inventories must deal with the complicating factor of bunker fuels, which are classified as international and currently not associated with a particular county, but may be regulated in future GHG-reduction treaties.

Due to limitations like those mentioned above, the development of alternative measurement-based approaches to quantifying CO_2 emissions must become a key area of focus for the segment of the scientific community which currently focuses on understanding natural CO_2 sources and sinks using techniques like inverse modeling. Developing these techniques to a more mature level and making use of currently available measurements (surface-based in situ, flask, aircraft and satellite) as well as those specific advances coming online within the timeframe targeted in this study (ICOS, OCO-2, etc.) will be a crucial component of the MRV activities.

Methods to Improve Emissions Inventory Estimates

Below are some examples that may be used to improve the uncertainties in the "bottom-up" emissions inventories. These include utilizing tracers of CO2 to better determine anthropogenic contributions from the natural global carbon cycle, further investigation of urban emissions, and development of flux estimates within a region to validate inventory estimates on a small geographic scale.

Measurements of Megacity CO_2 Emissions

Urban emissions dominate anthropogenic CO_2 emissions globally, accounting for more than 70% of the total anthropogenic contribution. In fact, CO_2 emissions from the 20 most populous megacities would represent the third highest "national" emissions level, trailing only emissions from China and the United States. Megacities contain the most concentrated area emissions, and provide us with excellent test cases, because of the high signal to noise and the relevance to treaty monitoring. Megacities are complex, multi-faceted regions, and there has been a paucity of measurements in these regions to contribute to the bottom-up inventories.

The types of cities to include in this study are megacities, industrial cities, high-technological cities, low-tech cities, quickly growing cities, well-established cities. This distribution of cities should give us a good idea of what kinds of emissions to expect in a given region.

A wide variety of measurements are required to properly attribute emissions and verify emissions inventories in urban centers. Measurements of $^{14}CO_2$ are the most proven method for quantifying the emissions contribution from fossil fuel combustion, but these measurements can only be made periodically from whole air samples, and require a difficult and expensive extraction process for analysis on a linear accelerator/mass spectrometer. Newman and coworkers have recently pioneered a continuous in situ method for accurate tracking of fossil fuel emissions in the Los Angeles megacity by determining CO(excess)/CO_2(excess) using simultaneous measurements within the LA urban dome and in a nearby pristine background environment. These measurements were validated against simultaneous $^{14}CO_2$ measurements during CALNEX. Measurements of saturated (ethane, propane) and unsaturated (benzene, acetylene) hydrocarbons, industrial halocarbons (HFC134a, CFC-12), etc. provide valuable correlative tracers for specific emissions processes and/or sectors, and may be used to differentiate emissions from different cities. Direct measurements of CH_4, N2O, and their isotopologues are required to verify total emissions for these much more uncertain GHGs. Remote sensing measurements of NO2 and SO2 have tremendous potential as tracers for emissions from industrial combustion.

There is an array of complementary measurement techniques that can be applied to quantify urban emissions. Protocols for surface in situ measurements in remote locations are mature, but significant research is required to determine the requirements for accurate surface in situ monitoring within the complex environment of an urban dome. Several vendors have recently produced accurate, moderate cost continuous in situ sensors with unprecedented precisions for specific trace gas targets (<0.1 ppm for CO_2; < 1 ppb for CH_4). Additionally, Griffith and coworkers have developed an operational Fourier-transform spectrometer solution that yields exceptional precisions for in situ measurements of more than 30 trace gases with integration times of ~5 minutes.

Vertically resolved profile measurements from aircraft and sondes (AirCore), as well as total column measurements from ground-based solar-viewing Fourier transform spectrometers (TCCON) are important for direct comparison with space-based measurements and for assessing the sensitivity of measured signals and inferred surface-atmosphere fluxes to transport.

Finally, the network design for measurements within an urban region must be sufficiently flexible and/or extensible that the network can adapt to new sources, changes in the urban landscape over time, and to take advantage of new information or measurement technologies.

The measurements must be coupled with mesoscale models (e.g. WRF) to assess the impact of transport within cities on source attribution. This may also help the data assimilation community assess the impacts of source patterns on their retrievals. We cannot fully rely on models, since we need to know wind profile and PBL height at a variety of locations, hence sondes and aircraft profiles are important (i.e. from profile information measured at airports or independently, if there are not enough airports or they are not well-distributed throughout the city).

Measurements of Doubly Substituted 638 CO_2 Isotopologue

There are numerous studies in which measured CO_2 isotopic fractionation – systematic enhancement or depletion of the abundance of a rare isotope such as ^{13}C or ^{18}O compared to a stochastically mixed sample of CO_2 generated from natural abundances of C and O – have been used to elucidate details of the CO_2 budget. For example, $\delta^{13}C$ is useful for discriminating marine from terrestrial photosynthesis and $\delta^{18}O$ can be used to distinguish terrestrial net photosynthesis from soil respiration.

The study of the extremely rare doubly substituted isotopologue $^{16}O^{13}C^{18}O$ (638) has been pioneered by Eiler and coworkers. The isotopic fractionation of 638, denoted Δ_{47}, can distinguish between fossil fuel combustion and biomass burning. Typical values of Δ_{47} range from 0.85 ‰ in clean air to 0.80 ‰ in polluted urban air [Affek and Eiler 2007][i].

Regional Fluxes

Due to the limitations in "bottom-up" self-reported emissions inventories, the KISS Monitoring Exchange of Carbon Dioxide workshop explored opportunities for "top-down" measurement of CO_2 and verification and validation of emissions estimates in current inventories. This section reviews possible space-based and in situ observational and modeling approaches necessary to improve our understanding of the exchange of CO_2 between the land, atmosphere, and ocean.

What is the state of regional flux observations and modeling?

Spatial and temporal distributions of CO_2 at the regional scale are typically determined by two classes of methods: 1) model extrapolation from small-scale flux observations or 2) inversion of atmospheric concentration data [Carouge, 2010]. An understanding of the current state of the science in these areas is necessary to assess the accuracy with which fossil fuel emissions can be attributed to specific geographic regions.

CO_2 flux observations include contributions from the natural carbon cycle in addition to anthropogenic components. An additional challenge in flux measurements is to determine the anthropogenic components. This attribution challenge is in addition to the overall flux measurement challenges such as sparseness and discontinuity of data and uncertainty in atmospheric models that are part of the data processing pathway. In few cases is it possible to directly measure only the anthropogenic CO_2 flux quantity directly.

Inez Fung's workshop presentation outlined a variety of ongoing ground and air based measurements with different spatial and temporal characteristics. Whole air flask samples collected at the surface by NOAA ESRL's Global Monitoring Division (GMD) Carbon Cycle Greenhouse Gases Group provide discrete measurements on a weekly basis; the site locations and measurement protocols are designed to measure background concentrations from large-scale footprints. Continuous in situ measurements at a smaller number of tall tower sites provide high frequency data, typically reported as hourly averages.

The areas represented by these measurements extend from 10 – 1000 km depending on the measurement height, the prevailing winds, and the local topography. Additional surface measurements come from instruments deployed on research ships, cruises of opportunity, and autonomous ocean platforms (buoys), which collect data with various frequencies.Satellites can provide valuable global measurements of column-averaged mole fractions of CO_2, although these have lower precision and accuracy than sub-orbital measurements.

Currently, there is no unified analysis framework in which all of these measurements are integrated and reconciled. Some of the challenges in integrating the observations include reconciling the different spatial scales, different observing frequencies, and different observing periods with varying meteorology and climate variability, as well as addressing incomplete representation of carbon cycle aspects (e.g. respiration may not be well represented). Additional challenges exist in the state of the models used to produce the data, especially in the atmospheric transport models and other forward models. Even with all of these observations, there are still large uncertainties around the carbon sources and sinks, further complicating the identification of fossil fuel contributions to the overall CO_2 emissions.

Evaluation of Regional Fluxes

Since CO_2 is a long-lived trace gas and the atmosphere rapidly mixes CO_2 emissions, anthropogenic CO_2 emitted at a specific place and time must be determined against large and also temporally varying background signal. The complicated interaction of local and global, natural and anthropogenic contributions to the CO_2 emissions problem has defeated attempts to achieve consensus on any unified international policy framework. It has also foiled all attempts to date to use existing global measurement networks to estimate accurate regional scale surface-atmosphere fluxes.

That said, relatively few countries and regions dominate the total global emissions. In fact, China and the United States together account for more than 40% of the global anthropogenic CO_2 emissions. Thus, research can focus on a small number of major source regions. This transforms the problem into one with potentially tractable technical and political solutions. Although it is essential to have broad international participation in any such policies to avoid efforts by major emitters to export emissions, for reasons described in more detail below, monitoring emissions from small source regions may be beyond the capabilities of current or near term global observational methods.

In addition to emissions from combustion of fossil fuel, other processes including modification of aerosol, methane, and land use have altered radiative forcing. The impact of these emissions, however, are either short-lived (aerosol) or are no longer major contributors to the change in total forcing (methane).

Pilot Project: Evaluate Regional Emissions from Global Observations

One high priority pilot project would be to evaluate the ability of global satellite observations to observe trends in regional scale emissions. The focus of this project would be: 1) to evaluate the total fossil-fuel derived emissions regionally and 2) to quantify the emissions resulting from tropical forest disturbance.

The goal for this effort is to detect changes in FF emissions of 10% from trend observations from the major source regions over a decade (e.g. 2012 – 2022). This goal has been selected since it is comparable to the emissions reduction targets in the Kyoto Protocol, as well as the magnitudes of change that have been advanced in several local and national policies. Given that emissions from China and India increased 8-10%/year during the 2000s, our goal seems quantitatively reasonable.

Notionally, we suggest the subdivision of source estimation as follows:

1. China
2. Continental US
3. India
4. EU-25
5. Russia
6. Australia
7. Japan
8. Tropical Forest Disturbance (primarily Brazil & Indonesia)

The deliverable will be an observational and analysis plan that can achieve such precision with a traceable and transparent approach.

The primary challenge in developing a monitoring system relying on atmospheric observations and top-down methods is signal to noise (or signal to clutter). Although the fossil emissions are large, the long lifetime of CO2 in the atmosphere dilutes the signals of these emissions. In addition, although the fossil emissions dominate the net flux to the atmosphere, gross fluxes of CO2 between the atmosphere and oceans and land dwarf the anthropogenic emissions producing noise or clutter.

There are many possible approaches that may allow the emissions of FF CO2 to be disentangled from those of the biosphere and oceans. For example, use of multiple tracers (e.g. 14C or combustion tracers such as CO, SO2, and NO2) may make improve the robustness of the ff estimation to error in our knowledge of the natural sources. Finally, any effort to develop methods to estimate ff emissions will certainly improve our knowledge of the natural carbon cycle and how it is being modified by the emissions of FF CO2.

The Attribution Challenge

Prather et al. [2009] identify the attribution of climate change to GHG emissions from a specific country as the primary challenge of GHG emissions monitoring. There are several aspects of current

measurement and analysis strategies that confound attempts at incontrovertible attribution to emissions from a specific nation or region. These are briefly summarized below.

Uncertainties in Transport Modeling

Identification of changes in CO_2 emissions from fossil fuel combustion (FFCO2) currently rely on transport models to relate observed CO_2 mixing ratios to upstream emissions. Detection and quantification of changes in FFCO2 will take place against a background of changing CO_2 driven by synoptic, seasonal, and interannual variations in transport superimposed on variable fluxes from the terrestrial biosphere and oceans. Note that variations in transport are often correlated with variations in vegetation and ocean fluxes that are driven by changing weather. For these reasons, modeled transport must be as realistic as possible to reduce errors in attribution of FFCO2 changes.

Transport models are not perfect and may have serious deficiencies in critical functions such as vertical convective transport and simulation of planetary boundary processes. We have very limited validation of the accuracy of tracer transport in the models near the Earth surface. To date, the range of transport uncertainty in global models has principally been approximated by using different transport model formulations and different wind fields [TransCom, Gurney et al., 2002]. The differences found have been large. This range may not capture the real world.

The error estimate in quantifying FFCO2 changes will require an evaluation of the transport error in the appropriate setting, i.e., for the specific study regions. Possible approaches to evaluating transport error include comparing multiple models at the same scale (e.g., global as TransCom), comparing global model transport to fine resolution models (ideally those/one with resolved convective transport), and comparison to diagnostic tracer observations.

Global transport models will be required for all cases, and these may be supplemented with regional models or higher resolution “zooms” for some domains. Transport with analyzed winds is required for direct comparison to observations on sub-decadal scales. Analysis periods of ~5-10 years will be likely needed to determine interannually varying FFCO2 trends with confidence.

In current inversion methods, the meteorological states used to drive transport models are mostly from reanalysis products, which were usually generated from a data assimilation system that used a model with different resolution and dynamics from transport models. The reanalysis data were then interpolated to be at the same resolution as transport models. Using reanalysis products to drive transport models can have the following consequences: 1) dynamical inconsistency between different variables because of interpolation; 2) interpolation error, which is difficult to quantify; 3) error in single meteorological state that is difficult to quantify. Generating ensemble meteorological states by assimilating meteorological observations simultaneously with atmospheric CO_2 data in the same transport model with and ensemble Kalman filter (EnKF) could overcome these problems, because it would no longer be necessary to interpolate the transport rates, and the ensemble meteorological

states generated through EnKF capture the joint uncertainty in the mean meteorological state and the atmospheric CO_2 state.

High precision observations of CO_2 in the middle atmosphere, such as those from the thermal infrared sounders TES [Chahine et al., 2008] and IASI [Crevoissier et al., 2009], lack sensitivity to surface fluxes [Chevallier et al., 2005], but may provide the constraints on atmospheric transport required to improve surface-atmosphere CO_2 flux estimates [Shia et al., 2006]. The upper tropospheric CO_2 seasonal cycle phases are reasonably well captured by the models such as GEOS-Chem and3-D MOZART-2, but the model simulations have smaller seasonal cycle amplitudes in the Southern Hemisphere compared with those in the Northern Hemisphere. Comparison of the simulated vertical profiles of CO_2 between data and the models reveals that the convection in the 3-D models is likely too weak in boreal winter and spring. Model sensitivity studies suggest that convection mass flux is important for the correct simulation of upper tropospheric CO_2. Therefore, high quality CO_2 measurements may be used to improve transport models [Jiang et al., 2008].

Aggregation Errors

Any signal in the atmosphere is a convolution of a space-time distribution of sources and sinks and the impact of transport and chemistry in the atmosphere. We seek to determine space-time integrals of these sources, usually but not always at the surface. Transport models must represent the sources and sinks on a finite grid. We may further choose to represent larger-scale patterns of sources and recover scaling factors for these. Kaminski et al., [2001] showed the inherent danger in this approach since errors in the small-scale patterns will be aliased into errors in the larger-scale estimates. This prompted the use of high-resolution studies where the patterns were left as free as possible. More recently, Bocquet (2005a,b) and Gerbig et al. (2008) showed the problems of this approach in which the information in the atmospheric signal is largely consumed in estimating the near-field contribution.

Clearly then there is a need to represent the small-scale variability of fluxes as well as possible without completely swamping the atmospheric signals in near-field contributions. This is achieved by providing the best possible information on small-scale structure, usually by using either models or proxy data. The attribution process is inherently statistical so also requires that all the uncertainties associated with these proxies or models must be rigorously included. since they play a part in the attribution,, the data on these proxies must fulfill the same requirements of openness and universality as all other parts of the data. We therefore recommend the generation of a coordinated database of proxies of economic activity. Such proxies are manifold but obviously include distributions of nightlights, power-plants, transportation networks, power grids and many others. Rather than forming an exhaustive list we should entrain the expertise of large-scale economic geographers and similar specialists.

Note added in proof: In addition to the Vulcan project [Gurney et al., 2009] providing high resolution emissions inventories for the US, there now exist several high-resolution global databases [Rayner et al., 2010; Andres et al., 2011; Oda et al., 2011]

Uncertainties in Fluxes from the Terrestrial Biosphere

As already noted, the joint challenge is to detect a change in patterns of flux then to attribute this change to FFCO2. This attribution is potentially confounded by other contributions to the detected signal. The problem does not arise from the strength of some confounding signal but rather its uncertainty. The problem differs greatly depending on the tracer used. The attractiveness of ^{14}C arises chiefly from its near direct link to fossil fuel fluxes [Turnbull et al., 2009]. For CO2 the main confounding variable (also called nuisance variables) is the flux due to net biospheric exchange. Direct estimates of this flux are so uncertain on large scales that it is usually estimated as the residual when all other terms have been considered [e.g. Cannadell et al., 2007]. Since there are no direct measurements at large scale, all estimates rely on models, either empirical [e.g. Reichstein et al., 2007 or mechanistic models (e.g. Krinner et al., 2005)]. Any estimate of uncertainty is based either on pointwise comparisons of these models with direct measurements [Chevallier et al., 2006] or estimates of model spread [Freidlingstein et al., 2006].

To quantify the confounding influence of uncertainty in biospheric flux on detection of emission trends we will perform a forward simulation using fluxes from a series of biospheric models. The quantitative question is whether the space-time signal of an emissions trend in a given region can be unambiguously detected among the background noise from the biosphere. The problem is identical to the univariate fingerprint detection method for climate change (e.g. Hegerl et al., 1997).

Uncertainties in Ocean-Atmosphere Fluxes

Large-scale monitoring of the global ocean carbon sink: The world's oceans play an important role in the global carbon cycle as they are a vast reservoir of carbon, rapidly exchange carbon with the atmosphere, and take up a substantial portion of anthropogenically released carbon from the atmosphere. Of the three major reservoirs with natural exchange rates fast enough to vary significantly on the timescale of decades to centuries (the atmosphere, terrestrial biosphere, and the oceans), approximately 90% of the carbon is located in the oceans. The oceans are able to hold much more carbon than the other reservoirs because most of the CO_2 that diffuses into the oceans reacts with seawater to form carbonic acid (H_2CO_3) and its dissociation products, bicarbonate (HCO_3^-) and carbonate (CO_3^{2-}) ions. The equilibration time scale for this exchange is about one year, so on a global scale surface water CO_2 generally increases at close to the same annual rate as the atmosphere [Takahashi et al. 2009]. On a finer scale, local physical or biological perturbation events can make surface water CO_2 significantly deviate from atmospheric equilibrium making it difficult to directly monitor changes in air-sea exchange with time.

Although ocean biology plays an integral role in the natural distribution of carbon in the ocean, there is no conclusive evidence that the ocean uptake and storage of anthropogenic carbon, thus far, involves anything other than a chemical and physical response to rising atmospheric CO_2. However, the ocean sink for carbon is variable in time [McKinley et al. 2004; Peylin et al. 2005, Canadell et al. 2007, LeQuere et al. 2009) and has the potential to become noticeably less efficient at absorbing carbon due to feedbacks with climate [Sabine and Tanhua, 2010]. There is a lot of uncertainty in these feedbacks and in the magnitude of their potential impact on the atmospheric pCO_2. However, it is clear that if the CO_2 sink in the ocean does decline in efficiency, additional reductions in anthropogenic emissions will have to be made in order to meet atmospheric pCO_2 targets. Thus, for policy purposes, we must be able to accurately monitor the ocean carbon sink through time.

The existing global oceanic carbon observatory network of repeat hydrographic surveys, time-series stations (ship-based and moored) and ship-based underway surface observations in the Atlantic, Pacific, and Indian Oceans provide a strong foundation of carbonate chemistry observations to begin constraining the ocean carbon flux of CO_2. However, enhancements are needed to accomplish the goals laid out here. Meeting the new observational constraints likely will involve implementing new technologies such as floats, drifters and autonomous vehicles to make additional observations more efficiently. The development of empirical algorithms relating in situ data to satellite products such as sea surface temperature, salinity, ocean color, sea surface height and surface winds will also allow interpolation of the data to global scales. Data assimilation (physical and biogeochemical) and inversion modeling studies will help delineate large-scale changes in water chemistry. Prognostic modeling will assist with attribution and prediction.

Although anthropogenic emissions are not typically associated with the oceans, providing a tight constraint on the ocean CO_2 fluxes can help with the delineation of sources and sinks on land.

Monitoring of the coastal ocean for constraining local emissions: For the smaller scale identification of local emissions sources, signals of variability and trends in the sink of carbon in the open oceans is unlikely to be distinguishable from signals of the terrestrial biosphere (D. Baker, personal communication 2010). However, the coastal oceans are likely to matter a great deal because their CO_2 fluxes are large and highly variable compared to the open ocean and they occur in close proximity to the land. These fluxes have the potential to "contaminate" both in situ and satellite-based observations purporting to capture terrestrial signals. Furthermore, coastal carbon fluxes are poorly understood from a mechanistic standpoint due largely to the paucity of data to constrain processes [Hales et al. 2008]. Finally, there are quantitatively important transfers of carbon from the land through groundwater, streams, rivers and estuaries into coastal waters [Cole et al. 2007, Tranvik et al. 2009]. This carbon is likely to be respired in the nearshore zone and returned to the atmosphere. There is some balancing of these effluxes slightly further offshore where nutrients delivered with the carbon begin to stimulate productivity, carbon uptake from the atmosphere, and export of carbon to the deeper waters and sediments [Cai et al., 2006; Chen and Borges, 2009]. However, the degree to which these fluxes balance

each other is unknown. It is clear that we must account for these transfers when quantifying carbon sources and sinks on land.

For coastal environments, a network of new hydrographic and ecological surveys will be required along with new coastal models. Meeting the observational constraints likely will involve implementing new technologies such as floats, drifters and autonomous vehicles to make additional observations more efficiently. The development of empirical algorithms relating in situ carbon data to physical parameters more frequently observed will assist extrapolation of the limited data sets in time and space. A series of flux towers in rivers, estuaries and coastal regions can help confirm estimated fluxes. Data assimilation (physical and biogeochemical) and inversion modeling studies will help delineate large-scale changes in water chemistry. Prognostic modeling will assist with attribution and prediction. Quantification of cross shelf water and carbon transports will require detailed water column measurements of carbon and biogeochemical parameters.

Example Pilot Projects

The examples below highlight projects that can be undertaken to quantify anthropogenic CO_2 emissions. The data and results from these studies would also provide the basis for assessing potential space-based monitoring solutions.

Commercial Aircraft Measurements OSSE

One cost-effective strategy that meets the criteria of coverage and openness is the instrumentation and automated reporting from commercial aircraft. This has been pioneered by the CONTRAIL program of Japan Airlines and is being further developed within the European IAGOS infrastructure <www.iagos.org>. To assess the potential value of expanding such a program one could perform an OSSE for the availability of daily vertical profiles over the world's collection of international airports. Although this will represent only a small fraction of the data returned by such a measurement programme, the large amount of data gained during cruising is unlikely to have strong impact on the determination of surface fluxes (e.g. Chevallier et al., 2010) but can act as a strong constraint on atmospheric transport. Even if only 1% of all commercial aircraft were instrumented, this data stream would revolutionize our insight into atmospheric CO_2 distributions.

Forward Modeling of Sensitivity to FFCO2 Changes

We will simulate CO_2 fields using general circulation models to identify the influence of fossil emissions on the total CO_2 field, which comprises fossil, oceanic, and biospheric CO_2. Presently, we have model results from the AM2 general circulation model in which total column CO_2 fields have been simulated. CASA biospheric fluxes, Takahashi ocean exchange, and fossil fluxes are used as boundary conditions. From the simulations, we identify neighboring regions with strong differences in column average CO_2 mixing ratio resulting from differential fossil fluxes. For instance, current uncertainty estimates for FFCO2 from China are in the range of 30% [Gregg et al., 2007]. We estimate that eastern China FFCO2 is 0.5-0.7 ppm higher in the annual mean than for western China. Although this difference must be

detected above a total column CO_2 dipole resulting from biospheric exchange that varies from -2.0 to 0.5 ppm CO_2 seasonally, the dipole over China provides a detectable signal from fossil emissions. As fossil emissions increase in eastern China, we predict this east-west dipole will scale with emissions, based on model output with doubled Chinese emissions.

In light of these model results, we propose further studying CO_2 fields from various transport models to generate influence functions from regional fossil fuel emissions. We will further study total column CO_2 fields, but will also sample the simulations to investigate the information to be gained from other observing platforms, such as space-based LIDAR satellites or unmanned aerial vehicles, which may provide vertically resolved CO_2 or provide the diurnal cycle in CO_2 as opposed to daytime only observations. These simulations will help us to pursue an enlightened observational strategy, as we expect the detectability of fossil fluxes to vary regionally and seasonally depending on the variability of natural CO_2.

Correlative Measurements

Correlative measurements of non-CO2 tracers plays important roles for improving CO2 flux estimates:

1. Identification of CO2 sources, e.g., biomass burning (CO), mobile (NO2), Coal fire plants (SO2)
2. Covariance with emissions
3. Constraints on transport
    a. CO, HDO

In the case of constraints on transport, correlative measurements play two important roles:

1. Mitigation of transport error
2. Improvement of transport forecast

As demonstrated by Liu et al. [2011], simultaneous assimilation of meteorological variables with CO_2 enables more accurate assessment of transport errors on the inferred CO_2 fluxes. This technique could be expanded to include multi-species assimilations (eg CO_2, CH_4, CO, NO2, SO2) to further leverage information in these data related to the emissions source.

Summary and Follow on Activities

The monitoring of anthropogenic CO_2 emissions is a national [Pacala et al., 2010] and international priority [UNFCCC, 1992]. This problem presents tremendous technical challenges since anthropogenic emissions must be disentangled from the much larger natural surface-atmosphere fluxes from the ocean and terrestrial biosphere. Despite these challenges, the workshop participants enumerated several solutions that could yield results within the next 5-10 years using existing methods and technology.

The most promising path forward appears to be monitoring anthropogenic emissions from megacities, as advocated by Pacala et al. [2010]. Approximately 35% of the total global anthropogenic CO_2 emissions

come from the world's 20 megacities (<1% of the total land surface area). The high concentration of emissions in such a small number of confined spatial domains leads to very favorable remote sensing conditions. In fact, satellite observations of megacity NO2 emissions [Zhang et al., 2007; Bierle et al., 2011] have demonstrated the feasibility of detecting signatures of fossil fuel combustion, and multi-year integrations of SCIAMACHY observations have shown persistently elevated CO_2 levels over major urban areas in Europe [Buchwitz, 2010]. Additionally, the establishment intensive ground-based CO_2 emissions monitoring in a few representative megacities would provide the benchmark data necessary to develop detailed models of urban emissions for designing optimized remote sensing solutions.

Accurate estimates of national level CO_2 emissions may also be undertaken, but required significant improvements in atmospheric transport modeling and inverse methods for successful results. The detection of anthropogenic emissions from the top 5-8 national emitters may be feasible with the OCO-2 satellite since anthropogenic emissions from these nations are largely constrained to the small fraction of the land area covered by densely populated urban centers. Preliminary calculations indicate that changes of ~10% in national level emissions may be discerned from ~5 years of observations. This sensitivity is commensurate with the changes required eg Kyoto Protocol emissions reductions policies. Such sensitivities should be able to clearly identify and quantify trends in anthropogenic CO_2 emissions from rapidly developing economies, such as China and India, where economic growth (and presumably CO_2 emissions) are expected to grow at the 7-10% per year level through 2020. The use of non-CO_2 observables (CO, NO2, SO2, etc.) in multi-species assimilations have the potential to significantly improve source attribution.

A major cultural challenge exists in convincing the MRV community of the value of objective validation of inventory-based emissions estimates by atmospheric concentration observations. Reconciling the top-down and bottom-up information will ultimately produce more robust inventory protocols and estimates (as in the excellent CO emissions inventories now in place in California), and point to new observations to minimize remaining uncertainties.

Opportunities exist to leverage activities proposed by the workshop participants with other projects being undertaken by national and international groups. The methodologies being developed under NIST sponsorship for detecting urban CO_2 emissions in the INFLUX experiment [Mays et al., 2009] can readily be transferred to larger cities in the US and elsewhere. The efforts defined in this report are well aligned with those that the Integrated Carbon Observing System (ICOS) is developing: a ground-based observing network and the analysis tools required to produce regular assessments of the GHG budget for Europe and surrounding regions. NASA has initiated a Carbon Monitoring System project to complement its continued development of satellite sensors for atmospheric CO_2 monitoring [NASA, 2010], and DOE recently commissioned a multi-national laboratory study to define the requirements for a greenhouse gas information system [Dimotakis et al., 2011].

Members of the workshop plan to submit a follow-on Large Study proposal to initiate sub-orbital Megacity CO_2 Monitoring and advanced remote sensing solutions focused on joint deployment of geostationary (GEO) and low earth orbit (LEO) satellites.

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